CLAIMS

1. Granular secondary particles of a lithium-manganese composite oxide which are granular secondary particles made up of aggregated crystalline primary particles of a lithium-manganese composite oxide, characterized in that

the granular secondary particles have many micrometer-size open voids therein, the open voids having an average diameter in the range of from 0.5 to 3 μm and the total volume of the open voids being in the range of from 3 to 20 vol.% on average based on the total volume of the granular secondary particles.

2. The granular secondary particles of a lithium-manganese composite oxide of claim 1, characterized in that the granular secondary particles have a specific surface area of from 0.2 to 1.0 m^2/g and an average diameter of from 5 to 30 μm, and the crystalline primary particles constituting the granular secondary particles have an average diameter of from 0.5 to 4.0 μm.

3. The granular secondary particles of a lithium-manganese composite oxide of claim 1, which are represented by the compositional formula $Li_XM_YMn_{3-X-Y}O_{4-Z}F_Z$ (wherein X, Y, and Z are such numbers that X = 1.0 to 1.2, Y = 0 to 0.3, and Z = 0 to 0.3; and M represents one or more elements selected from Al, Co, Ni, Cr, Fe, and Mg).

4. The granular secondary particles of a lithium-manganese composite oxide of claim 1, characterized in that the content of one or more boric acid compounds contained as an impurity in the granular secondary particles of a lithium-manganese composite oxide is lower than 0.0005 in terms of molar ratio between the manganese and boron (B/Mn) contained in the lithium-manganese composite oxide.

5. The granular secondary particles of a lithium-manganese composite oxide of claim 4, characterized in that the boric acid compounds contained as an impurity are lithium borate and/or lithium sodium borate.

6. A process for producing the granular secondary particles of a lithium-manganese composite oxide of claim 1, characterized by comprising spray-drying a slurry prepared by dispersing a fine powder of a manganese oxide and a fine powder of lithium carbonate or by dispersing a fine powder of a manganese oxide, a fine powder of lithium carbonate, and a compound containing the element M as described in claim 3 to thereby granulate the slurry and then calcining the granules at a temperature of from 700 to 900°C.

7. The process for producing granular secondary particles of a lithium-manganese composite oxide of claim 6, wherein the fine powder of a manganese oxide and the fine powder of lithium carbonate have an average particle diameter of 1 μm or smaller.

8. A process for producing the granular secondary

particles of a lithium-manganese composite oxide of claim 1, characterized by comprising spray-drying a slurry prepared by dispersing a fine powder of a manganese oxide, a lithium source, and an agent for open-void formation or by dispersing a fine powder of a manganese oxide, a fine powder of lithium carbonate, a compound containing the element M as described in claim 3, and an agent for open-void formation to thereby granulate the slurry and then calcining the granules at a temperature of from 700 to 900°C.

9. The process for producing granular secondary particles of a lithium-manganese composite oxide of claim 8, characterized in that the agent for open-void formation is a substance which has an average particle diameter of 1 μm or smaller and disappears upon heating.

10. The process for producing a lithium-manganese composite oxide powder of claim 6, characterized in that a compound which is a compound of an element other than manganese, lithium, fluorine, and the M described in claim 3 and is not an agent for open-void formation is added as an additive to the slurry.

11. The process for producing granular secondary particles of a lithium-manganese composite oxide of claim 10, characterized in that the additive is a boron compound, and that the compound is added to the slurry in an amount in the range of from 0.0005 to 0.05 in terms of molar ratio

between manganese and boron (B/Mn) and, after the calcining, the boron is removed by water washing to such a degree that the molar ratio (B/Mn) decreases to below 0.0005.

12. A non-aqueous electrolyte secondary battery characterized by employing the lithium-manganese composite oxide powder of claim 1 as a positive active material.